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Polychromatic liquid crystal laser arrays towards display applications

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Abstract: Band-edge liquid crystal lasers are of interest for a number of applications including laser projection displays. Herein, we demonstrate simultaneous red-green-blue lasing from a single liquid crystal sample by creating a two-dimensional laser array fabricated from dye-doped chiral nematic liquid crystals. By forming a pitch gradient across the cell, and optically pumping the sample using a lenslet array, a polychromatic laser array can be observed consisting simultaneously of red-green-blue colors. Specifically, the two-dimensional polychromatic array could be used to produce a laser-based display, with low speckle and wide color gamut, whereby no complex fabrication procedure is required to generate the individual 'pixels'.

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OCIS codes: (140.3290) Laser arrays; (120.2040) Displays; (230.3720) Liquid-crystal devices; (160.5293) Photonic bandgap materials.

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1. Introduction

Laser-based displays offer a number of advantages over their lamp-projection counterparts [1]. These include a wider color gamut (almost three times wider than conventional displays), higher resolution and contrast ratio, and excellent color saturation. The coherence of laser-based display systems also facilitates opportunities for holographic video projection [2]. Moreover, unlike lamp-projectors, the depth of focus is unlimited and the projection onto non-plane surfaces is trivial. Both of these benefits are a result of the fact that the laser beams are collinear; a feature that is not present in the lamp projectors. Developments in recent years have focused on using red-green-blue (R-G-B) lasers. Further, in order to access all the chromaticity values present on a CIE1931 (Commission Internationale de l'Éclairage) diagram, which are permissible within the boundaries set by the wavelengths of the three lasers lines, electro-optic modulators are used to vary the brightness of each laser.

In order for laser displays to become widespread, the integration of semiconductor technology or a suitable alternative is highly desirable. A number of reports have produced working prototypes using diode-pumped solid-state lasers [3] or vertical cavity surface emitting lasers (VCSELs) [4]. One drawback with semiconductor lasers, however, is that discrete devices containing different materials are needed thus demanding that each emitter is fabricated separately and then combined to form a display unit. The solid-state/OPO combination, despite its broad wavelength range, requires additional optics and techniques such as sum frequency mixing to generate the R-G-B output. Consequently, the identification of a single material system as a monolithic full color emitter is intensely sought after as this would facilitate a reduction in the system dimensions as a whole not to mention simplifying the fabrication process.

The liquid crystal (LC) laser exhibits many characteristics which make it an attractive alternative to semiconductor lasers. First and foremost, in contrast to semiconductor lasers, any wavelength can be chosen ranging from the deep ultraviolet to the near infra-red by adjusting the position of the photonic band gap (PBG) through chemical, mechanical, or electrical stimuli, whilst at the same time choosing a suitable light harvester for the desired wavelength range. The undesirable property of speckle can also be reduced through the use of short pulse lengths, or polydomain samples generating broader line-widths [5]. Typical output energies range between hundreds of nJ/pulse up to 10 μ J/pulse, whilst slope efficiencies are in the range of 1 – 30 % [6]. Several different incarnations of the LC laser have been reported in the literature but it should be stressed that whilst this paper reports on recent developments associated with the band-edge laser [7-9], the same principles of enhanced operation may also be applied to other LC laser device structures.

Successful operation of an LC laser requires that the energy density of the pump beam exceeds the minimum threshold for lasing. For a typical pump laser delivering tens of μ J/pulse, this corresponds to spot diameters in the range of 5 – 100 μ m. Increasing the pump energy density will increase LC laser output, but at high levels will eventually lead to detrimental effects such as optical orientation of the LC and also dye bleaching. Recently [5], we have shown that by using a 10 \times 10 lenslet array one can distribute the pump energy across the entire LC cell, and therefore create a two-dimensional array of LC lasers. Optical

reorientation and other high energy density side-effects can therefore be substantially reduced. Furthermore, assuming sufficient energy density within each individual spot, the active gain region of the LC is also substantially increased. These individual lasers then recombine to form a single output at distances greater than ~5 cm from the sample (or alternatively can be recombined using condensing optics), enabling high power outputs to be achieved. A similar approach has previously been applied to VCSELs to increase the output power, although for VCSELs more complicated fabrication procedures are required.

Current studies of LC lasers are typically conducted using *Q*-switched solid-state lasers, such as an Nd:YAG laser, as the pump source. The short pulses (ps or ns) ensure reduced speckle, and due to the high slope efficiencies, large peak powers can be obtained. In addition, studies [10-13] have shown that a range of wavelengths from a single chiral nematic (N*LC) sample can be obtained by using more than one laser dye and forming a pitch gradient across the cell. A broad band of lasing wavelengths are then individually obtainable by translating the sample relative to the pump beam. As a result, LC lasers are an attractive alternative as the full color emitter in a laser projection display. However, in order to realize the full potential of these lasers simultaneous R-G-B lasing from a single pump source is required. By combining gradient pitch LC samples with array-based pumping techniques, this paper reports that it is possible to achieve a multi-colored laser array consisting of red, green, and blue emissions when photo-pumped at a single fixed pump wavelength which is achieved without using complex fabrication procedures. In fact, R-G-B lasing from a liquid crystal defect-mode laser has recently been demonstrated [14, 15] using the innovative approach of stacking single-pitched polymer films to form a Fibonacci sequence. Using an alternative approach, we show that by combining gradient pitch LC samples with array-based pumping techniques, it is possible to achieve a multi-colored laser array simultaneously consisting of red, green, and blue emissions when photo-pumped at a single fixed pump wavelength. This is achieved without using complex fabrication procedures.

2. Sample preparation

In selecting the dyes for this study, absorbance and fluorescence measurements were carried out using dye-doped nematic samples. Several dyes were tested for their suitability for emission in the red, green, and blue. In each instance, a low concentration of dye (~1 wt %) was dispersed into the nematogen mixture BL093 (Merck KGaA). Each one was doped into the LC host and then capillary filled into anti-parallel rubbed 5 μm cells after mixing in a bake oven. Results for the three most suitable dyes i.e. DCM, Coumarin 540A, and Coumarin 504, are presented in this study.

The transition dipole moments of the dyes used in this study were found to have positive order parameters in the LC hosts. As a result, these mixtures exhibit preferential lasing at the long-wavelength photonic band edge. This is due to the greater overlap between the polarization vector of the electromagnetic wave and the transition dipole moment of the dye [16]. Consequently, the laser wavelength (λ) is related to the pitch of the LC (p) and the 'local' refractive index parallel to the director (n_{\parallel}) through the expression $\lambda = n_{\parallel} p$. We therefore control the pitch with the concentration of chiral dopant in the LC mixture.

The pitch can also be affected by temperature [17, 18]. All samples reported here were designed to operate at a single fixed temperature of 25 $^{\circ}\text{C}$, but it should be noted that a variation in this temperature of 10 $^{\circ}\text{C}$, for these mixtures, corresponds to a change in emission wavelength of only approximately 5 nm. Thermal control or compensation could be implemented to reduce this further in any practical device, if required.

For the laser samples, three dye-doped chiral nematic mixtures were prepared. These samples are referred to herein as the Red, Green, and Blue laser samples. For the Red sample, 5 wt % of the high twisting power chiral additive BDH1305 (Merck KGaA) was dispersed into the commercially available nematogen mixture BL093 (Merck KGaA) to generate a photonic band gap that had a long wavelength edge at 610 nm at 25 $^{\circ}\text{C}$. To this mixture was added ~1 wt % of the laser dye DCM (Lambda Physik). Due to the shorter pitch required, the

Green laser sample consisted of a higher concentration of BDH1305 (~6 wt %) in BL093 which resulted in a band gap with a long-wavelength edge at 530 nm at 25 °C. For emission in the green wavelength range ~1 wt % of the dye C540A (Exciton) was dissolved into the host mixture. Finally, the Blue laser sample comprised of 7 wt % BDH1305 and 1.7 wt % C504 (Exciton) in BL093. The resulting mixture created a N*LC with a long-wavelength band-edge at ~470 nm at 25 °C. Each mixture was placed into a bake oven overnight at a temperature of 100 °C to ensure thermal mixing of the constituents before being capillary filled into glass cells. The LC cells were constructed from two pieces of 1.1 mm thick glass, coated with anti-parallel rubbed polyimide alignment layers. Substrates were spaced apart by 10 μ m beads and glued together to form an empty cell. This thickness was chosen because it corresponds to the region of maximum slope efficiency, as determined experimentally in previous work [6]. Once cells were filled, optical microscopy was used to confirm that the LC texture was uniform and the solubility of the dye in the LC host was good.

3. Experimental

Absorption and emission spectra were recorded on an Olympus polarizing microscope that was connected to a USB2000 spectrometer. White light from the Halogen bulb was used to obtain the absorption profiles whilst the emission from a mercury lamp mounted on a reflection arm was employed to generate fluorescence from the samples. In both cases, samples consisting of only BL093 and the dye were used for these measurements to avoid the modification in the spectra which would result from the presence of the band-gap. Transmission spectra of the Red, Green and Blue laser samples as well as optical micrographs of the textures of the pitch gradient cells were also recorded on the microscope set-up.

The pumping module for the Red, Green, Blue lasers was in the form of a combination of a solid-state laser (Spitlight, Innolas) and an Optical Parametric Oscillator (OPO). Repetition rates and pulse widths were 2 Hz and 5 ns, respectively, delivering 40 μ J/pulse. The pump light from the OPO (430 nm) had a Gaussian profile and was incident upon a lenslet array, which spatially divided the beam up into 100 parts. The 10 \times 10 lenslet array used in this experiment was from Suss Microoptics and has a 1.015 mm pitch and a 28 mm radius of curvature. The plano-convex lens array is described in detail elsewhere [5]. Pump light passing through the lens-array is focused onto the LC cell thus generating 100 separate active gain regions at the sample. Each region then gives rise to a laser output provided the energy density is above the threshold. To capture the near-field emission profile, a 4 \times microscope objective was positioned after the sample so as to collect and magnify the near-field two-dimensional array so that it could be viewed clearly on a white screen a distance of 50 cm from the sample. Photographs of the screen were captured using a Canon 350D digital single lens reflex (SLR) camera. Emission spectra of either the recombined (far-field) array or of individual sources within the array were recorded using a USB2000 (Ocean Optics) spectrometer which had a resolution of 1.3 nm. The screen was replaced with a focusing lens, which coupled the laser emission into an optical fiber leading to the spectrometer. All measurements were carried out at 25 °C, to ensure that our designed mixtures lased at the required wavelengths, and so that thermally induced pitch (and emission wavelength) changes between samples and experiments were negligible.

4. Results and discussion

4.1 Red/green/blue laser arrays

Figure 1 shows the absorption and emission spectrum of each dye dispersed into the nematic host BL093 along with the transmission spectrum for white light for the Red, Green and Blue laser samples. Plotted on the primary axes of Figs. 1(a), 1(c) and 1(e) are the absorption bands, whereas the fluorescence spectra are plotted on the secondary axes. The absorption peaks of all three dyes lie within close proximity of one another and are the reason these dyes were chosen: to enable pumping at a single wavelength by the OPO module. The dashed line indicates the wavelength of the pump beam relative to the absorption maximum. Order

parameters of the transition dipole moments of the dyes (S_T) were found to be $S_T = 0.28$ (DCM), $S_T = 0.19$ (C540A), and $S_T = 0.36$ (C504).

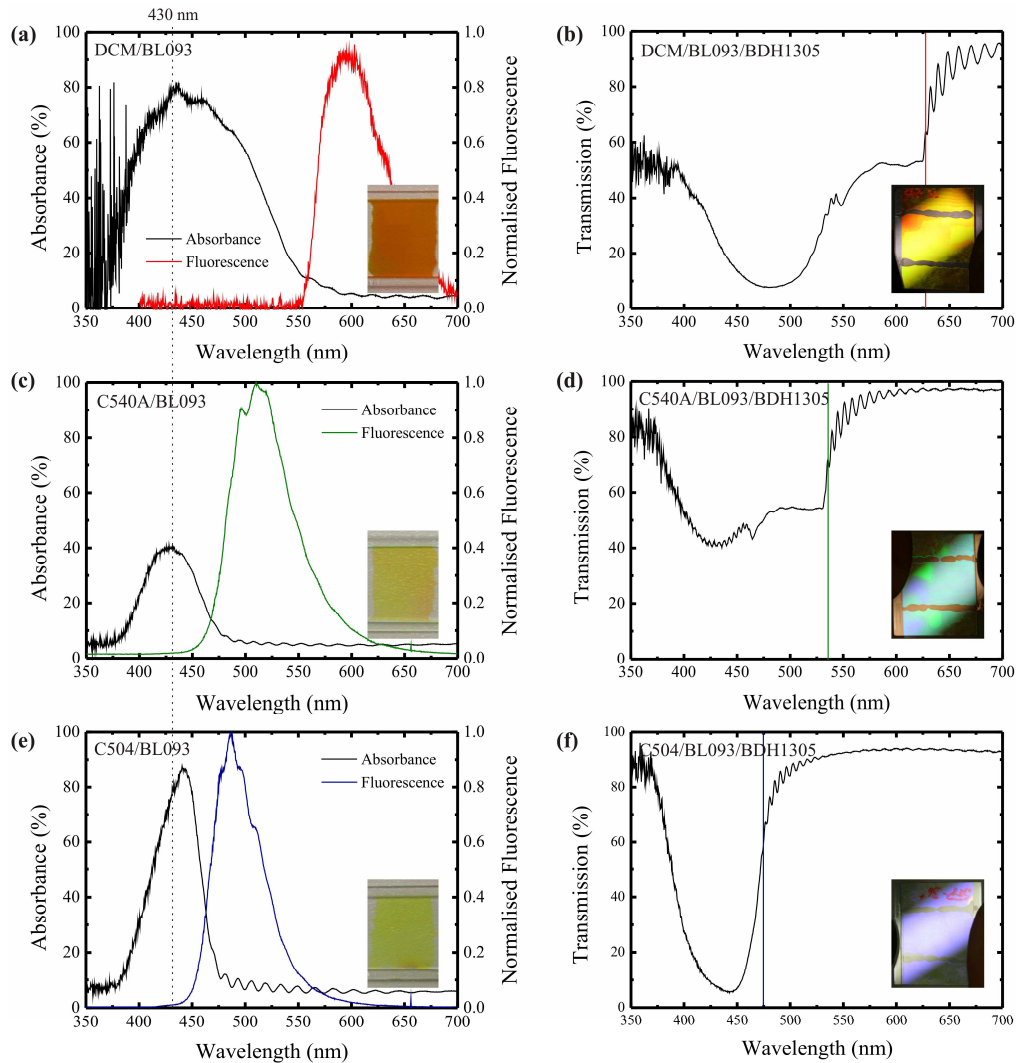


Fig. 1. Absorbance and fluorescence spectra (left) of three different dyes; (a) DCM, (c) C540A and (e) C504, mixed in a nematic liquid crystal (BL093) host, showing fluorescence emission peaks in the red, green and blue regions of the spectrum respectively, whilst sharing a common absorbance at around 430 nm (inset images illustrate the cells' appearances due to absorbance of the dyes). The graphs on the right, (b), (d) and (f) show the corresponding transmission spectra of the same three dyes in their chiral hosts (BL093 with BDH1305), illustrating the resulting photonic band gaps (and inset, selective reflection from the cell). The corresponding long band-edge positions are highlighted, showing where lasing can occur in the red, green and blue regions of the spectrum.

It is known that in a liquid crystal host PM597 is a more efficient emitter than DCM [6]; however, its absorption band is much narrower than DCM and cannot be optically excited at wavelengths as short as 430 nm. Fluorescence maximums for the three dyes were found to occur at 600 nm, 525 nm, and 490 nm for DCM, C540A and C504, respectively. As a result, the long-wavelength band-edges were chemically tuned (through control of chiral dopant concentration) to align within close proximity with these wavelengths. However, in order to

widen the color gamut, the Red and Blue samples were tuned to wavelengths of ~620 nm and 470 nm, respectively, whilst still remaining at the higher regions of the gain curve. At the bottom right hand corners of these figures are photographs of the cell showing absorption of the cells as recorded by a CCD camera for white light illumination. The similar appearance of both C540A and C504 is in accordance with the absorption spectra; the more orange appearance of DCM is due to the broader absorption band.

Figures 1(b), 1(d), and 1(f), are transmission spectra for the three laser samples. For each of the plots it can be seen that the absorption of the dye covers the short wavelength regions, in accordance with the absorption spectra shown in Figs. 1(a), 1(c) and 1(e), and that the band-gap for the Red and Green samples is present at longer wavelengths. The band-gap is not visible for the Blue sample due to the fact that the desired laser wavelength is very close to the long-wavelength side of the absorption band (Fig. 1(e)). It is shown that the long wavelength edges are at 620 nm, 530 nm, and 470 nm (indicated by the solid colored lines) for the Red, Green, and Blue samples, respectively. Also included in the figures are photographs of the cells (bottom right hand corner) depicting the selective reflectance of each sample. The reflected colors in these images represent the sum of all wavelengths within the photonic band gap and thus appear slightly bluer in color than the resulting chromaticity of the corresponding LC laser.

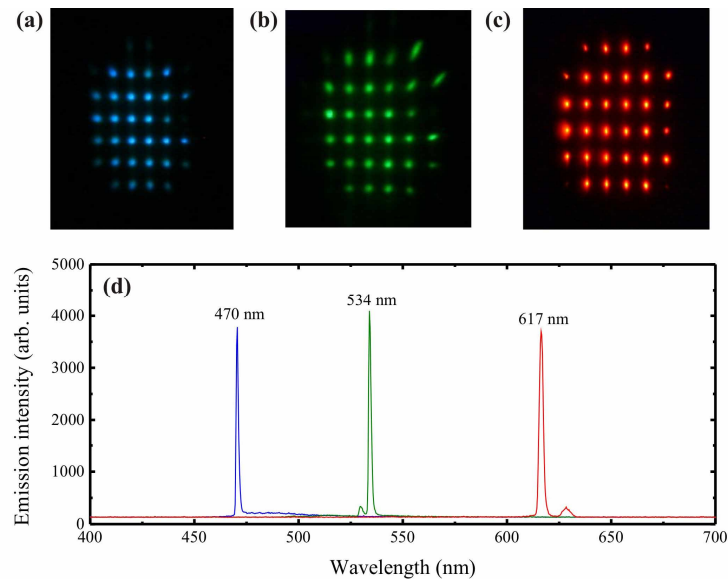


Fig. 2. (a), (b) and (c) Monochromatic LC laser array emissions in the blue (470 nm, C504), green (534 nm, C540A) and red (617 nm, DCM) respectively, and (d) the corresponding emission spectra (taken by recombining the array in the far field).

When optically excited at 430 nm the three samples are found to emit monochromatic laser lines at 617 nm (Red), 534 nm (Green), and 470 nm (Blue). Figure 2 presents three photographs of the near-field two-dimensional array when magnified and viewed in the far-field using a 4 \times microscope objective. The laser array is a 10 \times 10 grid with spacing between nearest neighbors of 1 mm. However, due to the numerical aperture of the objective some of the outer spots of the array are not collected and a maximum of 32 spots is recorded at any one time. Emission spectra for the lasers are shown in Fig. 2(d) when the array is recombined into a single spot in the far-field. Line-widths are less than 1 nm and are limited by the resolution of the spectrometer. The monochromatic output is a good indication that across the entire array the sample is a single monodomain with little variation in the pitch of the helix.

The benefit of an array-based pumping approach is that it enables much higher total energies to be delivered to the sample without degradation of the structure as a whole. Studies

have shown [6] that at specific input energies (for a single pump spot) the output energy reaches a saturation limit and a further increase results in a decrease in energy. For conventional laser systems this is known as thermal roll-over. Using a phase-locked LC laser array (i.e. zero phase-shift between neighboring members) is therefore of interest for scaling up the output energy of LC lasers with good spectral properties, provided the laser wavelength remains within the gain maximum of the dye.

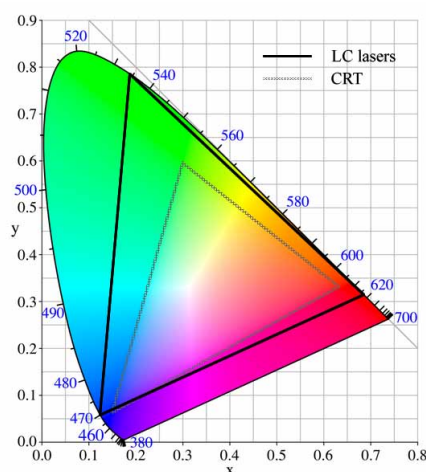


Fig. 3. A CIE1931 chromaticity space diagram. The inner triangle represents the color gamut from a typical CRT display, whilst the outer triangle represents the far greater potential performance from a liquid crystal laser display using three laser sources operating at 470 nm, 534 nm and 617 nm respectively.

The color gamut that would be available when combining these three lasers is shown on the CIE1931 diagram presented in Fig. 3. The horse-shoe colored area represents the full range of chromaticities visible to the human eye. The grey triangle corresponds to the outer boundaries of the color gamut obtained using the phosphors of a typical cathode-ray tube (CRT). A similar triangle (darker line) can also be drawn for the three LC lasers described above, linking points near the outer edge of the diagram at monochromatic laser wavelengths of 470, 534 and 617 nm. It can be seen that the resulting bounded area and corresponding color gamut is appreciably larger using these LC lasers in comparison to a conventional CRT, which would give rise to a display with greatly improved color resolution. Further improvements could also potentially be made by engineering the green laser to have a wavelength of closer to approximately 520 nm (thus generating a triangle of yet greater area). Alternatively, the low cost and ease of fabrication of LC laser cells allows the practical possibility of increasing the number of component colors from three to four (or more).

4.2 Polychromatic laser arrays

The three previously discussed laser arrays, shown in Fig. 2, correspond to three separate samples. In order to generate a single material system that emits R-G-B (or more colors) simultaneously we combine the array pumping approach with a pitch gradient cell. An illustration of the principle of the pumping/emission arrangement is presented in Fig. 4. Using the lens array, 430 nm light from the OPO is focused onto the cell whereby each column of the lens array photo-excites a separate domain of the cell which has its own predefined pitch. By using the appropriate dyes for a given pitch, multiple laser wavelengths are emitted simultaneously.

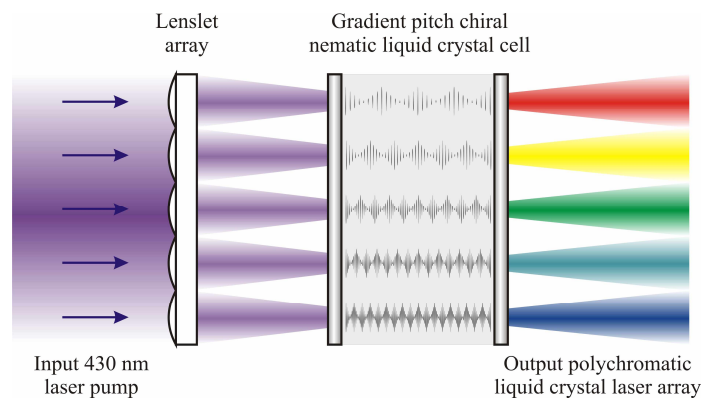


Fig. 4. Diagrammatic representation of a polychromatic liquid crystal laser array. Pump light is focussed, using an array of lenslets, onto a gradient pitch chiral nematic liquid crystal cell (doped with appropriate laser dyes). Multiple wavelength laser emissions are emitted simultaneously from different regions from within the same cell.

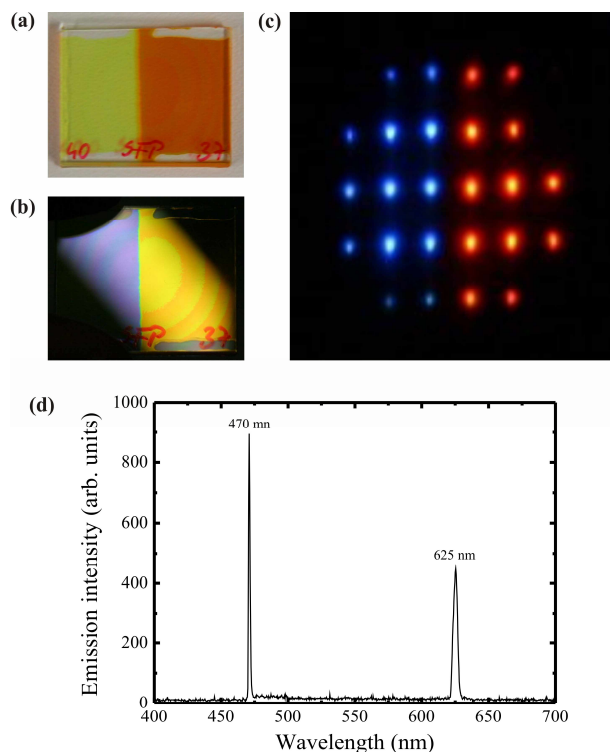


Fig. 5. Images of the gradient pitch liquid crystal laser cell, made by filling the cell with two different liquid crystal and dye mixtures from opposite sides, immediately after filling, where diffusion in the cell has yet to occur. Photographs of the cell's characteristic (a) absorbance in ambient light conditions, and (b) selective reflection. When pumped at 430 nm using a lenslet array, lasing occurs in the cell: (c) is an image of liquid crystal laser emission from the cell, and (d) the corresponding emission spectrum, illustrating two distinct simultaneously occurring single-mode laser wavelengths.

To create the pitch gradient sample, custom cells were fabricated which consisted of gaps in the glue at the midpoint of the cell in order to allow air to escape into side chambers during

filling. The blue and red laser samples were filled simultaneously into opposite sides of the cell, forming a distinct step-boundary where the two samples meet. A photograph of the cell immediately after filling is shown in Fig. 5(a). In order to obtain emission in the green, 0.5 wt% of C540A was dispersed into the Red laser sample prior to filling. Figure 5 also includes a picture of the cell in reflectance (Fig. 5(b)) and the near-field array across the boundary (Fig. 5(c)). All images were taken within 1 hour after filling of the cell. The array consists of columns of red and blue laser spots at 625 nm and 470 nm, respectively. The well-defined columns of red and blue indicate that the sample consists of two large monodomains. This is reinforced by the emission spectrum for the recombined spot, which shows two monochromatic lines at 470 nm and 625 nm. The red emission is slightly longer than that shown in Fig. 2 due to the fact that the pitch has been diluted by a small amount with the addition of the green dye (C540A).

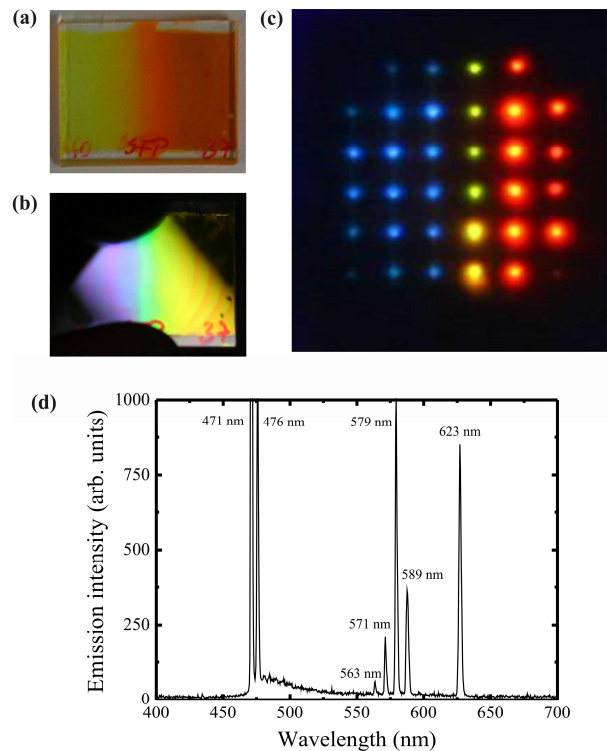


Fig. 6. Images of the gradient pitch liquid crystal laser cell, made by filling the cell with two different liquid crystal and dye mixtures from opposite sides, five days after filling, where diffusion in the cell has begun to occur. Photographs of the cell's characteristic (a) absorbance in ambient light conditions, and (b) selective reflection. When pumped at 430 nm using a lenslet array, lasing occurs in the cell: (c) is an image of liquid crystal laser emission from the cell, and (d) the corresponding emission spectrum, illustrating simultaneously occurring polychromatic laser emission.

Five days after filling and the characteristics of the cell have changed as a result of both diffusion of the dyes and the formation of a pitch gradient, leading to simultaneous polychromatic emission. Results are presented in Fig. 6 where it is evident that from the photographs of the cell (Figs. 6(a) and 6(b)) that the boundary is now less well defined. Figure 6(a) demonstrates absorption in the cell under white light illumination and how this varies across the cell where the dyes have diffused together. Figure 6(b) shows, through selective reflection, the subtle change in pitch across the boundary as the photonic band-gap and the selectively reflected light varies gradually from blue-green-red. When the sample is now optically excited at 430 nm the LC laser array consists of columns of blue and red as before

but now is separated by a column of green (563 nm) and yellow spots (589 nm). The recombined spectrum for the overall array is shown in Fig. 6(d). In total, seven laser lines are present (although greater or fewer lines could be made visible by simply replacing the lens array with one of a finer or greater pitch); each line is separated by more than 5 nm from its nearest neighbor. There is still a line at ~470 nm and ~625 nm but additional lines occur at 563 nm, 571 nm, 579 nm, and 589 nm. Despite the fact that the C540A dye was added to the Red laser sample, no emission is observed around the region of 530 nm. Alternative samples were prepared whereby the C540A was dissolved into the Blue laser sample only and then in both the Red and the Blue samples, but in all cases no emission was observed in the 530 nm region of the electromagnetic spectrum. This is believed to be due to Forster transfer between the C540A and DCM dye since the fluorescence band of C540A overlaps the absorption band of DCM. In order to cover all three wavelengths without using or encountering Forster transfer, one possible solution is to divide the cell into separate compartments. Such an approach is currently under investigation.

The non-uniformity of the laser peaks are due to the difference in efficiencies at different wavelengths of the laser dyes being used. Some laser peaks will also be brighter due to certain single-pitch domains within the cell sharing a larger proportion of the pump beam. One must also take into account the weakening intensity of the Gaussian-shaped pump beam (and therefore of the resulting laser emission) towards the outer edges of the aperture.

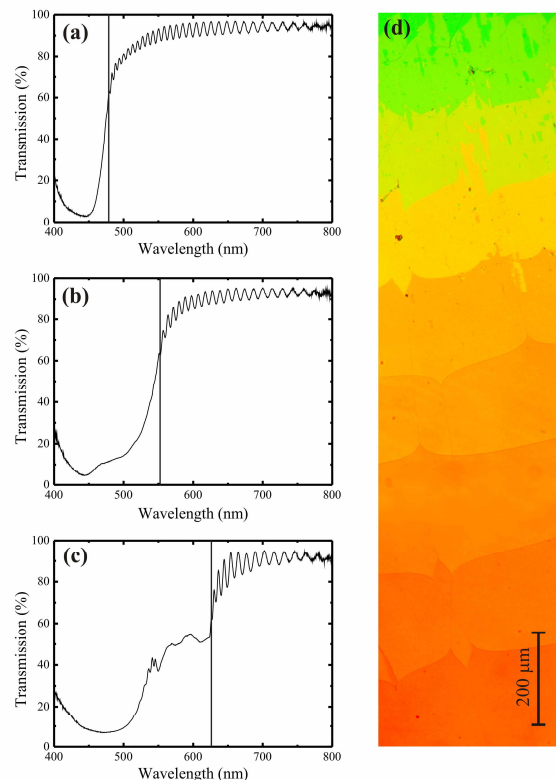


Fig.7. Transmission spectra of different spatially positioned sections within the gradient pitch cell, showing changes in the position of the photonic band edge from (a) blue, to (b) green, to (c) red. A polarized microscopy image of the gradient pitch cell (d) shows characteristic banding of colors, where discrete jumps in the chiral pitch occur to preserve an integer number of molecular rotations between cell walls.

Figure 7 shows the transmission spectra of the cell for three different spatial positions across the boundary corresponding to red (620 nm), green (550 m), and blue (470 nm) emission, indicated by the solid lines. To the right of the graphs is an optical micrograph of

the sample texture between crossed polarizers. The characteristic bands of color represent the regions of different natural pitch which are forced to adopt a half-integer number of turns between the antiparallel aligned boundaries of the cell. This pitch gradient, which occurs in discrete jumps, results from the competition of the forces between the elastic free-energy associated with the natural pitch of the helix and the surface anchoring forces.

In addition to display applications, these results have another distinct benefit. By observing the subtle change in wavelength across the x-y plane it is possible to build up a 2D map of the chiral nematic liquid crystal structure itself, tracking how the pitch topology changes with time. This approach could be applied to structures such as cholesteric elastomers to help build up a picture of a structure that includes a vast number of polydomains, and to observe how the structure changes under mechanical deformation.

After a long period of time, the sample diffuses such that the extreme wavelengths (470 – 660 nm) are separated by almost the entire spatial separation of the cell (i.e. over a distance of 2 cm). Although this is undesirable for a practical device it can be easily resolved by using smaller LC cells, or by using polymer stabilization to restrict further diffusion within the structure. In actual fact, a recent report has shown that such a structure can be easily stabilized using photo-polymerization of chiral nematic liquid crystals consisting of monoacrylate and diacrylate components to prohibit further diffusion with time [19]. To some extent, the time-dependent diffusion can be taken advantage of by fixing at a desired point in time to stabilize the structure corresponding to a particular set of emission characteristics. Furthermore, through techniques such as UV initiated phase separation of polymer and liquid crystal [20], one could construct arbitrary divisions or compartments within a gradient pitch cell, isolating regions of desired laser emission, and preventing further diffusion from occurring.

5. Conclusions

In this paper, we have presented a method of achieving simultaneous red-green-blue laser emission from a liquid crystal band-edge laser when optically excited with a single visible wavelength. This is achieved by using a combination of a lenslet array, generating a 10×10 array of focused spots at the sample, and a pitch gradient LC cell. It is shown that the near-field LC laser array consists of well-defined columns of red-green-blue outputs. Such an approach offers a facile alternative to laser arrays based upon semiconductors and potentially provides a simple route towards the fabrication of liquid crystal lasers for laser projection displays, with low speckle and wide color gamut.

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